

Coherent control of ac Stark allowed transition in Λ system

Gennady A. Koganov* and Reuben Shuker†
 Physics Department, Ben Gurion University of the Negev
 P.O.Box 563, Beer Sheva 84105, Israel

We show that quantum-interference-related phenomena, such as electromagnetically induced transparency, gain without inversion and enhanced refractive index may occur on electric-dipole forbidden transitions. Gain/dispersion characteristics of such transitions strongly depend upon the relative phase between the driving and probe fields. Unlike allowed transitions, gain/absorption behavior of forbidden transitions exhibit antisymmetric feature on the Rabi sidebands. Absorption/gain spectra possess extremely narrow sub-natural resonances.

PACS numbers: 42.50.Gy, 42.62.Fi, 32.70.Jz

Quantum-interference-related phenomena, such as electromagnetically induced transparency (EIT), lasing without inversion (LWI) etc., are based on the interference between two independent quantum channels [1, 2, 3, 4, 5, 6, 7, 8, 9, 10]. Traditional treatment of such phenomena typically involves a three level scheme and two coherent fields, a strong driving field and a weak probe one, applied to two dipole-allowed transitions, followed by measurement of the absorption and the dispersion of the probe transition.

In this Letter we show that dipole-forbidden transitions can also exhibit amplification, enhanced dispersion and other coherent phenomena. The electric field component of the driving laser field breaks the space spherical symmetry and renders the parity not well defined, as in *dc* Stark effect. In other words, the presence of a strong driving field exerts *ac* Stark effect and thus breaks the spherical symmetry of the system and creates an infinite ladder of dressed states [11, 12]. An interesting signature of the forbidden transitions is the antisymmetric character of the gain and dispersion on the Rabi sidebands. We found that gain and dispersion properties of such transitions are phase sensitive as they strongly depend upon the relative phase between the driving and probe fields. Our results open a perspective for new type of phase sensitive spectroscopy in a wide spectral range.

We consider a three level scheme in Λ -configuration shown in Fig. 1. A strong driving field with Rabi frequency $\Omega_L \exp(i\varphi_L)$ and a weak probe field with Rabi frequency $\Omega_P \exp(i\varphi_P)$ are applied to atomic transitions $|b\rangle \rightarrow |c\rangle$ and $|a\rangle \rightarrow |c\rangle$, respectively. Note that the phases φ_L and φ_P are introduced explicitly in the driving and the probe fields, respectively.

Due to selection rules the transition $|a\rangle \rightarrow |b\rangle$ is dipole-forbidden by parity which is well defined in the absence of the laser fields. However, the parity becomes ill defined due to the presence of the *ac* electric fields of the two impinging lasers. These fields break the space symmetry and mix states of different parity. Hence the originally

forbidden transition becomes undefined. In this sense it becomes *dynamically* allowed. We call such dynamically allowed transitions *ac*-Stark allowed (ACSA) transitions. In other words the interaction with two coherent fields, at least one of which is strong, creates an infinite ladder of dressed states [11, 12], so that transitions between the dressed states are not necessarily constrained by the selection rules for a free atomic system. In a sense this is a different variant than the usual EIT or LWI schemes as here the transition is forbidden to begin with and there is no issue of population inversion. The transition is dynamically allowed solely due to the quantum interference. The system Hamiltonian in the interaction picture and

the rotating wave approximation is given by

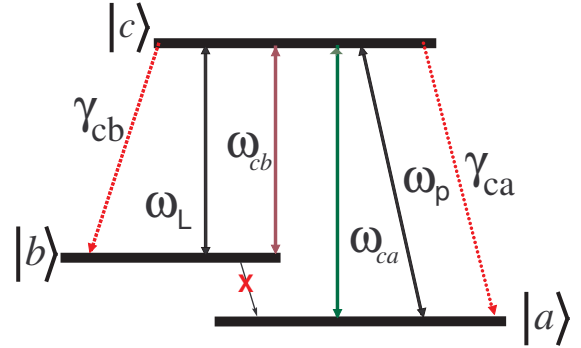


FIG. 1: Schematic Λ three level configuration. Transition $|a\rangle \rightarrow |b\rangle$ is initially dipole-forbidden (marked with red cross).

$$H = \begin{pmatrix} 0 & \Omega_L e^{i\varphi_L} & \Omega_P e^{i\varphi_P} \\ \Omega_L^* e^{-i\varphi_L} & \Delta_L & 0 \\ \Omega_P^* e^{-i\varphi_P} & 0 & \Delta_P \end{pmatrix} \quad (1)$$

where Ω_P and Ω_L are Rabi frequencies of the probe and the driving fields, respectively, and Δ_P and Δ_L are corresponding detunings. Abbreviated manifold of dressed states created by the strong driving field Ω_L is shown in Fig. 2. As will be seen in the following, maximal gain is achieved when the probe and the drive lasers

*Electronic address: quant@bgu.ac.il

†Electronic address: shuker@bgu.ac.il

are in "dressed" two-photon resonance with transitions between the dressed states marked with thick arrows (red and orange on line), i.e. at $\Delta_P - \Delta_L = \pm R$, where $R = \sqrt{\Omega_P^2 + \Omega_L^2}$. The corresponding semiclassical dressed states in the case of resonant drive field are given by

$$|\pm\rangle = \frac{1}{\sqrt{2}} \left(\frac{\Omega_P}{\sqrt{\Omega_P^2 + \Omega_L^2}} |a\rangle + \frac{e^{i\Delta\varphi}\Omega_L}{\sqrt{\Omega_P^2 + \Omega_L^2}} |b\rangle \pm e^{-i\varphi_P} |c\rangle \right) \quad (2)$$

These dressed states are superposition of states of different parity and hence are not constrained by the selection rules for atomic states $|a\rangle$, $|b\rangle$, and $|c\rangle$.

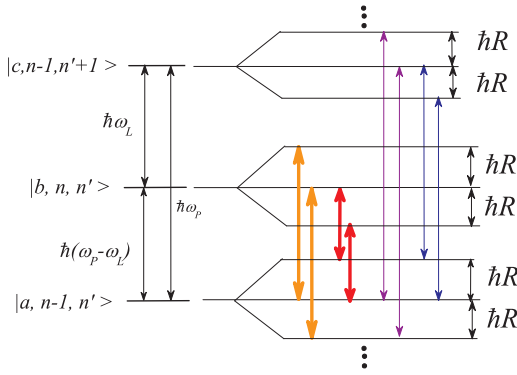


FIG. 2: Dressed states picture at bare two-photon resonance $\Delta_P - \Delta_L = 0$. On the left: manifold of bare states labeled by the atomic level index a , b , or c , the driving field photon number n , and the probe field photon number n' . On the right: dressed states of coupled atom+field system. Strong phase sensitive gain without inversion is obtained on the ACSA transition $|a\rangle \rightarrow |b\rangle$ at frequencies $\omega_P - \omega_L + R$, left couple of thick arrows (orange on line) and $\omega_P - \omega_L - R$, right couple of thick arrows (red on line), see also Fig. 3(c). Two couples of thin arrows (blue and violet on line), show the probe transition frequencies $\omega_P - R$ and $\omega_P + R$, where gain is also possible. $R = \sqrt{\Omega_P^2 + \Omega_L^2}$ is the generalized Rabi frequency.

We have solved analytically master equation for the atomic density matrix ρ with the Hamiltonian (1) in steady state and calculated the coherences ρ_{ac} and ρ_{ab} on the probe and the ACSA transitions, whose imaginary and real parts are related to absorption/gain and dispersion, respectively. The main features can be qualitatively understood from the approximate expressions for the coherences ρ_{ac} and ρ_{ab} on probe and ACSA transitions at small probe field $\Omega_P \ll \Omega_L$, although all graphical results presented in the following have been obtained from the exact analytic solution of the steady state master equation. Approximate formulas for the coherences are given by

$$\rho_{ac} = - \frac{2i(\Delta_L - \Delta_P)\Omega_P e^{i\varphi_P}}{(\gamma_{ca} + \gamma_{cb})(\Delta_L - \Delta_P) + 2i(\Delta_P(\Delta_L - \Delta_P) + \Omega_L^2)} \quad (3)$$

$$\rho_{ab} = \frac{2i\Omega_P\Omega_L e^{i(\varphi_P - \varphi_L)}}{(\gamma_{ca} + \gamma_{cb})(\Delta_L - \Delta_P) - 2i(\Delta_P(\Delta_L - \Delta_P) + \Omega_L^2)} \quad (4)$$

Here γ_{ca} and γ_{cb} are spontaneous decay rates from the upper state $|c\rangle$ to states $|a\rangle$ and $|b\rangle$, respectively, $\Delta_P = \omega_{ac} - \omega_P$ and $\Delta_L = \omega_{bc} - \omega_L$ are one-photon detunings between the laser frequencies ω_P and ω_L and the corresponding atomic frequencies ω_{ac} and ω_{bc} . The two-photon detuning $\Delta_P - \Delta_L = \omega_{ab} - (\omega_P - \omega_L)$ contains the frequency ω_{ab} of originally dipole-forbidden atomic transition $|a\rangle \rightarrow |b\rangle$ indicating possible oscillations at that frequency. It is important to note that due to the presence of the exponent factor $\exp[i(\varphi_P - \varphi_L)]$ in the numerator of Eq. (4), the coherence ρ_{ab} is *phase sensitive*. Varying the relative phase $\Delta\varphi = \varphi_P - \varphi_L$ between the probe and the drive fields allows for an interchange between absorptive and dispersive line shapes of $|a\rangle \rightarrow |b\rangle$ transition, defined by imaginary and real parts of the coherence ρ_{ab} , respectively. It is instructive to consider two particular cases of "bare" and "dressed" two-photon resonances, when $\Delta_P = \Delta_L$ and $\Delta_L - \Delta_P = \pm\Omega_L$. At $\Delta_P = \Delta_L$ ("bare two-photon resonance") the coherences become $\rho_{ac} = 0$ and $\rho_{ab} = -(\Omega_P/\Omega_L)\exp(i\Delta\varphi)$, so that for transition $|a\rangle \rightarrow |b\rangle$ both absorption and dispersion can take small values between $-\Omega_P/\Omega_L$ and Ω_P/Ω_L , depending on the relative phase $\Delta\varphi$. This accentuates the importance of the phase shift between the two laser fields and the phase sensitivity of the ACSA transition. The absorption ρ_{ac} on the probe transition $|a\rangle \rightarrow |c\rangle$ equals to zero, the signature of EIT and dark state[1, 2].

A different picture is obtained when the probe field is tuned in resonance with transitions between dressed states, i.e. when the two photon detuning equals the Rabi frequency of the driving field ("dressed two-photon resonance"). The dressed resonance condition and the coherences are given by

$$\Delta_L - \Delta_P = \pm\Omega_L \quad (5)$$

$$\rho_{ac} = - \frac{2i\Omega_P}{\gamma_{cb} + \gamma_{ca} + 2i\Delta_L} e^{i\varphi_P} \quad (6)$$

$$\rho_{ab} = \pm \frac{2i\Omega_P e^{i\Delta\varphi}}{\gamma_{cb} + \gamma_{ca} - 2i\Delta_L} \quad (7)$$

In this case both ρ_{ac} and ρ_{ab} are not small since they are proportional to the probe Rabi frequency Ω_P rather than to the ratio Ω_P/Ω_L between the probe and the driving field Rabi frequencies, as in the above mentioned bare resonance case. Also gain and dispersion on the ACSA transition are not symmetric with respect to the bare two-photon resonance $\Delta_P = \Delta_L$, as evident from Eq. (7). In particular, at zero phase shift $\Delta\varphi = 0$ and zero

drive detuning $\Delta_L = 0$ there is strong gain with anomalous dispersion at $\Delta_P = -\Omega_L$ and strong absorption with normal dispersion at $\Delta_P = \Omega_L$. Changing the phase shift to $\Delta\varphi = \pi$ results in an opposite sign of ρ_{ab} , so that there is absorption with normal dispersion at $\Delta_P = -\Omega_L$ and gain with anomalous dispersion at $\Delta_P = \Omega_L$.

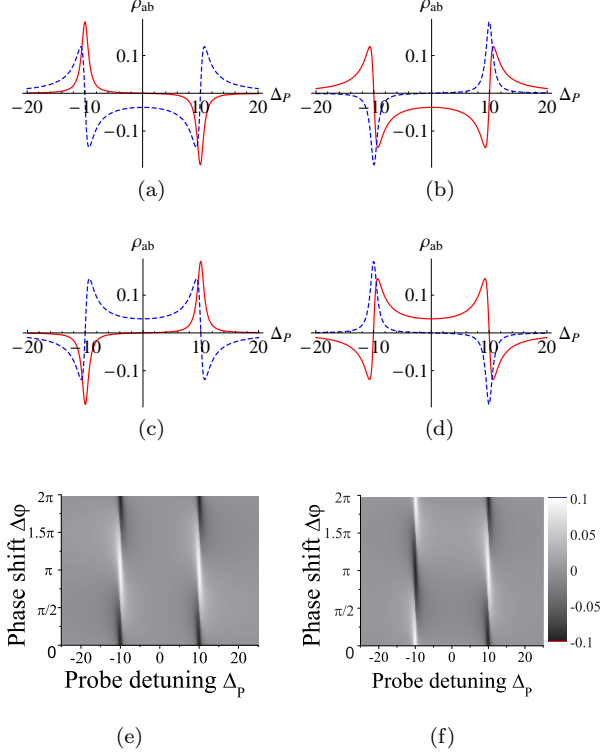


FIG. 3: $\text{Im}[\rho_{ab}]$ (solid curve, red on-line) and $\text{Re}[\rho_{ab}]$ (dashed curve, blue on-line) as functions of probe detuning at (a) $\Delta\varphi = 0$, (b) $\Delta\varphi = \pi/2$, (c) $\Delta\varphi = \pi$, and (d) $\Delta\varphi = 3\pi/2$. The antisymmetric behavior of the gain is accompanied by symmetric behavior of the dispersion at specific phase shifts, and vice versa. Parts (e) and (f) show contour plots of steady-state coherences ρ_{ac} and ρ_{ab} at probe and ACSA transitions, respectively. White/black color corresponds to maximal gain/absorption. Parameters: $\gamma_{ca} = \gamma_{cb}$, $\Omega_L = 10\gamma_{cb}$, $\Omega_P = 0.37\gamma_{cb}$.

To get a quantitative picture, exact analytical solution for the steady state coherence ρ_{ab} is drawn in Fig. 3 as a function of probe detuning Δ_P for various values of $\Delta\varphi$. When the probe and the driving fields are *in-phase* ($\Delta\varphi = 0$, see Fig. 3a) there is a strong gain with negative dispersion slope at $\Delta_P = -\Omega_L$, and absorption with normal dispersion at $\Delta_P = \Omega_L$. Inverse picture is obtained when the probe and driving fields are *out of phase*, i.e. $\Delta\varphi = \pi$ (see Fig. 3c). If the field phases are $\pm\pi/2$ -shifted (see Figs. 3b and 3d), the absorption profile takes a dispersive shape while the dispersion behaves in absorptive-like manner - an indication of the quantum interference. To compare absorption/gain properties of the probe and ACSA transitions contour plots of imaginary parts of ρ_{ac} and ρ_{ab} are shown in Figs. 3e and

3f, respectively. One can clearly see the difference: the probe coherence ρ_{ac} is symmetric with respect to bare two-photon resonance $\Delta_P = \Delta_L$ so that there is either absorption or gain simultaneously on both side-bands $\Delta_P - \Delta_L = \pm\Omega_L$ (two white traces at $\Delta\varphi = \pi$ and two black traces at $\Delta\varphi = 0(2\pi)$), while the coherence ρ_{ab} on the ACSA transition is antisymmetric with respect to the origin, so that there is always gain on one side-band and absorption on the other (white trace on one side-band is always accompanied by the black trace on the opposite side-band). The frequency at which amplification takes place can be tuned by varying the driving field intensity because maximal gain is obtained at $\Delta_P - \Delta_L = \pm\Omega_L$.

Another intriguing manifestation of quantum interference is obtained when the probe field is not weak with respect to the driving one. Figure 4 demonstrates four essentially different cases controlled by the *phase shift* between the probe and driving fields: (i) reduced refraction index at $\Delta\varphi = 0$ (fields in-phase, Fig. 4(a)), (ii) strong absorption with normal dispersion at $\Delta\varphi = \pi/2$ (Fig. 4(b)), (iii) strongly enhanced refraction index at $\Delta\varphi = \pi$ (out-of-phase fields, Fig. 4(c)), and (iv) strong gain without inversion and anomalous dispersion at $\Delta\varphi = -\pi/2$ (Fig. 4(d)). The results shown in Figs. 4(c) and 4(d) are especially important as they hint for two interesting potential applications: laser or/and optical amplifier with a wide spectral range of operation (see Fig. 4(d)), and a controllable atomic dispersion in wide spectral range (see Fig. 4(c)). Moreover, as one can observe from 3D plot of the imaginary part of the coherence ρ_{ab} (see Fig. 4(e)), the unique broad flat-top gain takes place in a wide range of phase shift $\Delta\varphi$ as well.

To calculate the absorption spectrum, the time dependent master equation has been solved numerically and then Fourier transform has been taken. Figure 5 plots the spectrum of the coherence ρ_{ab} for various values of the probe detuning Δ_P . As one can see, in addition to two resonances at Rabi-shifted transition frequencies $\omega_{ab} \pm \Omega_L$, a very sharp peak appears at $\omega_{ab} + \Delta_P - \Delta_L$. This peak becomes especially strong by a few orders of magnitude, at the "dressed" two-photon resonance, i.e. at $\Delta_P - \Delta_L = \pm\Omega_L$ (see Fig. 5c). Interestingly, the peaks have extremely narrow line widths (see insets in Figs. 5c and 5d). We have found numerically that increasing accuracy of digital Fourier transform gives rise to vanishing line width, i.e. ideally, if one could perform Fourier transform with infinite accuracy, the peaks would look like δ -function. This can be explained by the absence of mechanisms of line-broadening on the forbidden transition in this model. Indeed, there are neither dissipative processes which could contribute to line width, nor the field broadening as no field is applied to this transition. These will, of course, give a finite line width.

Sharp strong resonance in the spectrum of $|a\rangle \rightarrow |b\rangle$ transition means that an efficient lasing is possible on this transition with extremely narrow spectral line. Moreover, such a laser would be tunable since its frequency can be controlled by varying both the frequency of either

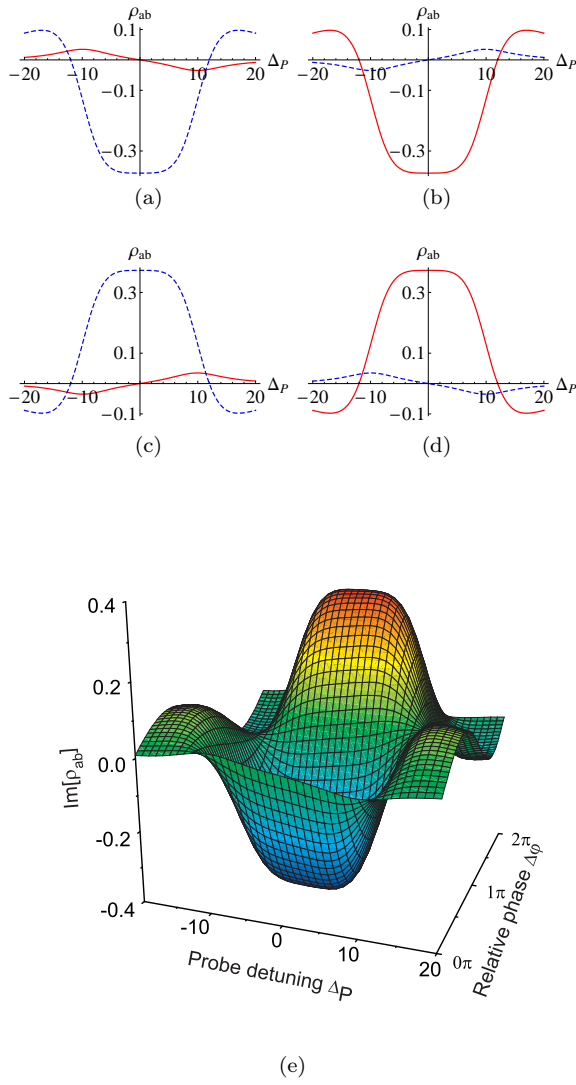


FIG. 4: Imaginary (solid curve, red on-line) and real (dashed curve, blue on-line) parts of the coherence ρ_{ab} on the ACSA transition at strong pump field as functions of probe detuning. Gain (absorption) is obtained at positive (negative) values. (a) Suppressed refraction index at $\Delta\varphi = 0$, (b) Strong absorption with normal dispersion at $\Delta\varphi = \pi/2$, (c) Enhanced refraction index at $\Delta\varphi = \pi$, (d) Gain without inversion with negative dispersion slope at $\Delta\varphi = -\pi/2$. Figure (e) shows 3D plot of imaginary part of ρ_{ab} as a function of the probe detuning Δ_P and of the phase shift $\Delta\varphi$. The dispersion behavior is similar and can be obtained by shifting the gain plot by $\pi/2$. Note the unique feature of flat-top gain and dispersion (not shown here) at particular values of the phase shift $\Delta\varphi$. Parameters: $\gamma_{ca} = \gamma_{cb}$, $\Omega_L = 10\gamma_{cb}$, $\Omega_P = 4.5\gamma_{cb}$.

the driving or the probe laser, and the intensity of the

driving laser.

It should be noted that our results are applicable to the systems where all three transitions are electric dipole allowed, such as Ruby and other solid materials [13], and semiconductor quantum wells [14].

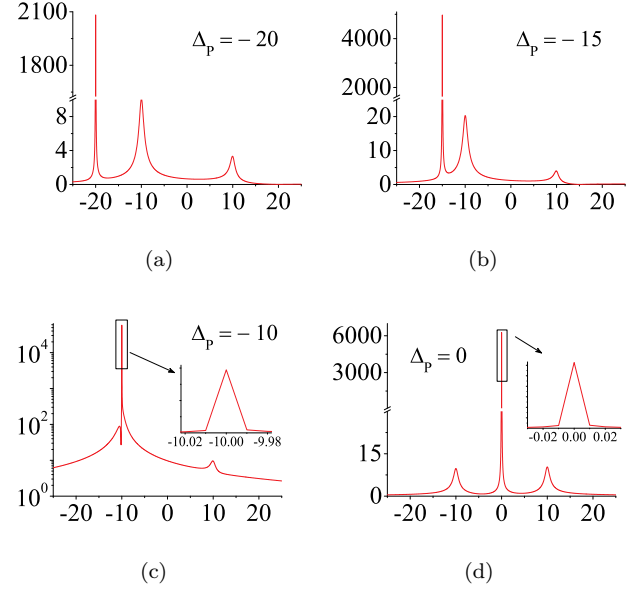


FIG. 5: Absorption spectrum of $|a\rangle \rightarrow |b\rangle$ transition (initially dipole-forbidden). Note sharp strong peak at dressed two-photon resonance $\Delta_P = \pm\Omega_L$ (see part c which is plotted in logarithmic scale). Parameters: $\Omega_L = 10$, $\Omega_P = 0.1$, $\Delta\varphi = 0$, $\Delta_L = 0$, $\Delta_P = -20$ (a), -15 (b), -10 (c), 0 (d).

Last but not least, similar results have been obtained for the ladder and the V schemes as well. In particular, strong gain without inversion can be obtained on dipole forbidden transition $|a\rangle \rightarrow |c\rangle$ of the ladder scheme (see Fig. 1c) which opens a new perspective for creating ultra-short wavelength and X-ray lasers without inversion. Details will be published elsewhere.

In summary, gain/absorption and dispersion characteristics of dipole-forbidden transitions in Λ scheme driven by two laser fields have been calculated. These are found to be strongly sensitive to the phase shift between the probe and the drive laser fields. It has been shown that $|a\rangle \rightarrow |b\rangle$ transition exhibits quantum-interference-related phenomena, such as EIT, gain without inversion, and enhanced dispersion. Absorption/gain spectra possess extremely narrow sub-natural resonances. At strong enough probe field both the gain and the dispersion exhibit a flat-top behavior controlled by the relative phase of the probe and the coupling laser fields.

[1] M.O. Scully, and M.S. Zubairy, *Quantum Optics*, Cambridge University Press, 1997.

[2] M. Fleischhauer, A. Imamoglu, and J.P. Marangos, Rev.

- Mod. Phys. **77**, 633 (2005).
- [3] G.S. Agarwal, Phys. Rev. A **55**, 2467 (1997).
 - [4] G.G. Padmabandu et al., Phys. Rev. Lett. **76**, 2053 (1996).
 - [5] J. Mompert and R. Corbalan, J. Opt. B: Quantum Semi-class. Opt. **2**, R7 (2000).
 - [6] A. Andre, M.D. Eisaman, R.L. Walsworth, A.S. Zibrov, and M.D. Lukin, J. Phys. B: At. Mol. Opt. Phys. **38**, S589 (2005).
 - [7] E. Arimondo, in *Progress in Optics XXXV*, edited by E. Wolf (Elsevier, Amsterdam) 1996, Chapt. V, pp. 258-354.
 - [8] A.S. Zibrov et al., Phys. Rev. Lett. **76**, 3935 (1996).
 - [9] J. Vanier, M.W. Levine, D. Janssen, and M.J. Delaney, IEEE-trans. Instrum. & Meas. **52**, 258 (2003).
 - [10] A. Godone, S. Micalizio, F. Levi, and C. Calosso, Phys. Rev. A **74**, 043401 (2006).
 - [11] C. Cohen-Tanudji, J. Dupont-Roc, and G. Grynberg, *Atom-Photon Interactions*, Wiley, New York, 1992.
 - [12] D. Braunstein and R. Shuker, Phys. Rev. A **64**, 053812 (2001), and references therein.
 - [13] E. Kuznetsova, Y. Rostovtsev, N.G. Kalugin, R. Kolesov, O. Kocharovskaya, and M.O. Scully, Phys. Rev. A **74**, 023819 (2006).
 - [14] M.D. Frogley, J.F. Dynes, M. Beck, J. Faist, and C.C. Phillips, Nature materials **5**, 175 (2006).